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Nonlinear Resonant Ultrasound Spectroscopy for Nondestructive Evaluation of Thermally Aged Small Pressed Pellets

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Abstract

As the high explosive Pentaerythritol Tetranitrate (PETN) ages, its microstructure changes in the form of microcracks, grain sintering, crystal coarsening, and via other mechanisms, which affects its explosive performance. However, current methods to interrogate the microstructure can be time consuming, hard to perform *in situ*, or do not necessarily produce meaningful information. Moreover, there is currently no proven benchscale method for characterizing the microstructure of pressed compacts, meaning this is a shortcoming for surveillance efforts. Nonlinear wave propagation in consolidated granular material, such as sandstones, concrete or in this case, pressed pellets, is a function of the microstructure and can be influenced by poor sintering of the grains, micro cracks and grain distribution. Nonlinear Resonant Ultrasound Spectroscopy (NRUS) is a tool that is able to measure the bulk hysteretic nonlinearity by resonating a sample at different amplitudes and observing the shift in resonant frequency. Typically, NRUS has been used on larger samples, but in this work, we use the technique to probe small pressed granular PETN pellets that have a diameter of 7.6 mm and a thickness of 1.2 mm that are artificially aged by placing in an oven for a period of time. The samples are resonated by adhering a small piezo-transducer and measured with a laser vibrometer. We found a positive correlation between the degree of nonlinearity and the amount of time artificially aging under heat treatment, indicating that NRUS is a promising tool for diagnosing damage in PETN.

Background and Research Objectives

The ability to predict and diagnose aging in PETN is of paramount interest to the detonator community. Only limited quantities of stockpile aged detonators are available for surveillance purposes to assess performance degradation. There is ample evidence that suggests the microstructure of PETN (Figure 1) is intimately connected with performance. Seminal works connecting PETN microstructure to explosives performance demonstrate that PETN Fisher Specific Surface Area (FSSA) decreases with time, and, in turn, decreasing FSSA correlates with increasing sensitivity and function time.² The PETN community has adopted FSSA as the benchmark metric for PETN aging. However, there are several problems associated with using FSSA as the metric for aging: (1) There are methods for “stabilizing” PETN that affect the rate at which a powder’s FSSA changes.³ Modern detonator grade PETN is stabilized, and as a result, recent artificial aging experiments show no change in FSSA.^{4,5} Contrarily, recent small angle neutron scattering (SANS) measurements on aged PETN indicate that the microstructure is indeed changing, and accelerated aging studies on detonators show indisputable performance

degradation.⁶ (2) FSSA is measured using the Fisher Sub-Sieve Sizer (FSSS), which is only capable of measuring powder FSSA. This precludes the use of the FSSS for stockpile-relevant PETN samples: pressed pellets confined inside detonator headers. Other research has inferred that powder density and confinement during aging affects the rate at which powder morphologies change.⁴

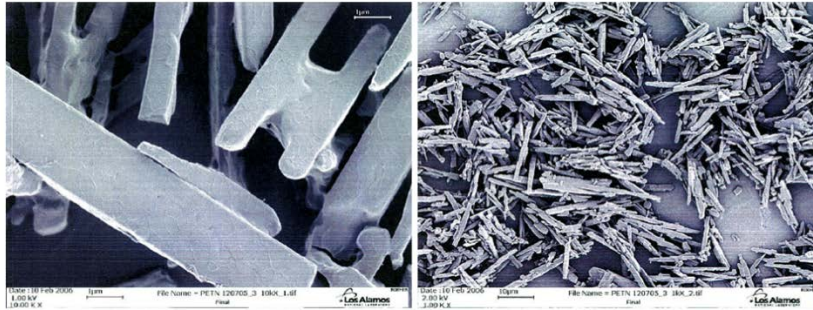


Figure 1: Micrographs depict Type 12 detonator grade PETN powder. Type 12 PETN is high surface area/small grain size powder. Crystallites have a large aspect ratio and are needle-like in shape.

The use of FSSA as a metric for aging in PETN implies that performance degradation can occur that we will not be able to detect. Further, we currently cannot characterize pellets that come out of the stockpile, and conclusions about their aging must be drawn from separate experiments. This indicates that our current metric for gauging the extent of aging damage in PETN, and more specifically in pressed PETN pellets, is inadequate.

The EVA Laboratory has pioneered a number of methods for material characterization, integrity, and nondestructive evaluation. A particular focus has been on nonlinear elastic wave techniques, in which wave propagation can depend on the strain amplitude, and a wave can become distorted as it travels. In particular, nonlinear characterization techniques have been demonstrated to be very sensitive in detecting differences in complex materials, for example, porous materials with many interfaces, micro cracks, and intricate geometry.⁷ A quantitative comparison performed in the EVA lab of crack density and nonlinear parameters was able to show a positive correlation, and furthermore, nonlinear signatures were sensitive to microscopic cracks long before the visualization threshold was met.⁸

One nonlinear diagnostic technique routinely used and studied on a variety of materials is Nonlinear Resonant Ultrasound Spectroscopy (NRUS). NRUS is able to make a bulk measurement of the degree of nonlinearity of the sample. The method works by resonating a sample at different amplitudes and observing the shift in the resonance frequency, which can be related to the nonlinear hysteretic term, α .

The goals of this project were threefold: (1) To determine if meaningful signal can be obtained using NRUS from such small pellets. Previous NRUS studies have typically been conducted on larger samples with different geometries. The challenge of small pellets is how to effectively excite them by adhering piezo sources to the surface. (2) To determine if the hysteric nonlinear parameter, α , obtained using NRUS can resolve differences between a pristine (not heat treated)

pellet and an artificially aged pellet. (3) To determine if α correlates well with explosive performance characteristics.

Scientific Approach and Results

Experimental Setup

PETN is an excellent candidate for nonlinear measurements such as NRUS since it exhibits many of the same properties as previous materials NRUS has been applied to (porous and fractured materials, *e.g.*: rocks, bone, concrete, and fatigued metals). PETN is initiated by a shock wave or rapidly expanding plasmas, and the sensitivity to initiation, function time, and other characteristics are affected by powder morphology.

To perform nonlinear measurements with NRUS a transducer oscillates the PETN sample through a range of frequencies while measuring the response with a laser vibrometer as shown in Figure 2. In this initial effort, epofix epoxy was chosen to adhere the transducers because of its ease of use and compatibility with PETN. A very thin layer of epofix was applied to the transducers which were then placed on the PETN and held down with a fixture pictured in Figure 2 and allowed to cure for one day. The transducers were piezo ceramic disks manufactured by STEMiNC (SMD07T02R412WL) with a diameter of 7 mm, thickness of 0.2 mm, and a resonant frequency of approximately 300 kHz. The response was measured using a Polytec class II laser vibrometer. Data was collected using a sophisticated data acquisition hardware and software system for performing NRUS, namely Resonance Inspection Techniques and Analyses (RITA)⁹.

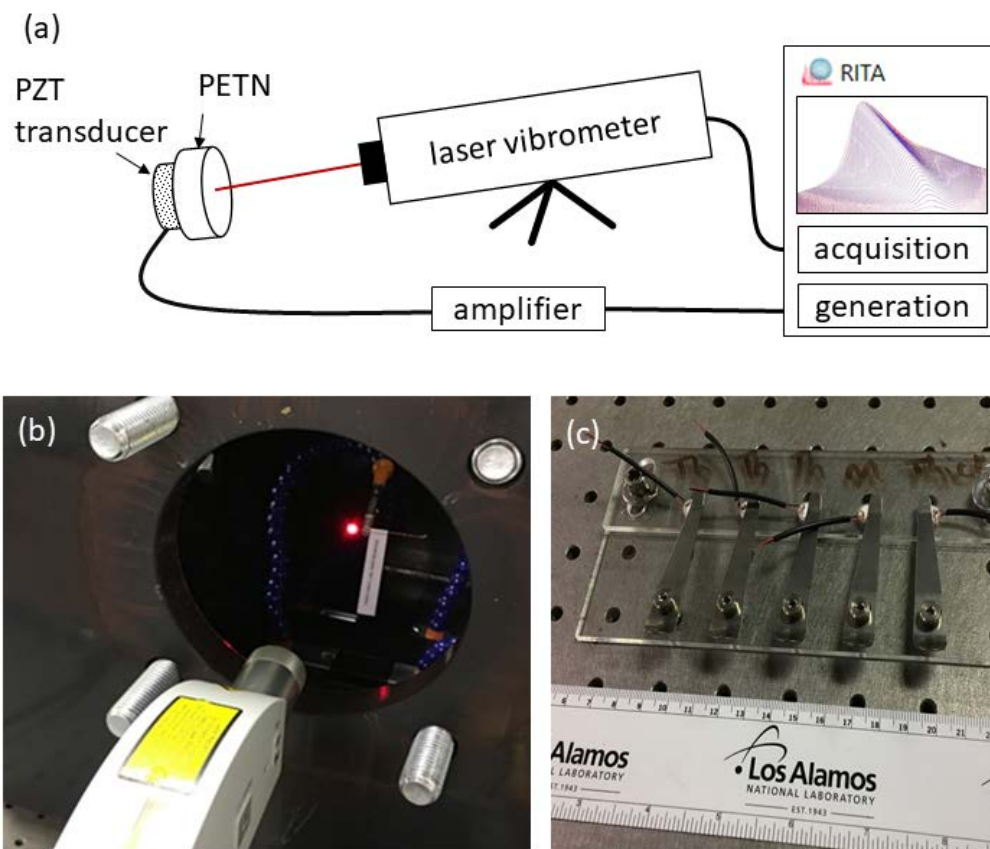


Figure 2. Experimental Setup: (a) schematic, (b) photograph of NRUS, and (c) photograph of epofix gluing holder.

The pellets were pressed to 1.65 g/cc and measured 1.6 mm tall and 7.6 mm in diameter. Forty pellets were kept pristine as a baseline for our measurements. And an additional forty pellets were aged according to standard artificial aging protocols by placing them in an oven at elevated temperatures (80°C, in this study) for predetermined lengths of time. The elevated temperatures increase the rate at which the sublimation/redeposition reaction occurs, which is accepted to be the dominant aging mechanism for PETN. Sublimation and redeposition of PETN is responsible for changes in the microstructure, including Ostwald Ripening and powder sintering. Ten pellets were removed from the oven after each the following durations: 4 weeks, 7 weeks, and 10 week. The pristine PETN pellets are “Ultra Pure White”. After 4 weeks aging at 80°C, the PETN pellets take on a “Palais White” hue. With 3 more weeks at 80°C, the pellets are a “hazelnut cream”¹⁰.

Results

An example resonance plot of PETN is shown in Figure 3. Peaks were chosen between 70 kHz and 120 kHz. As the driving amplitude is increased, the resonance frequency shifts lower, indicating a softening effect. The change in resonance frequency is plotted against measured

peak velocity in Figure 3. At low amplitudes, the shift in frequency is small and the linear regime is exhibited; however, as the amplitude increases further, the slope also increases as the nonlinear regime is reached. To find the nonlinear hysteretic coefficient, α , which is defined as the slope in the nonlinear regime, the slope of this data was calculated for a series of ten consecutive amplitudes along the entire data set, and then the maximum slope was used as the measurement.

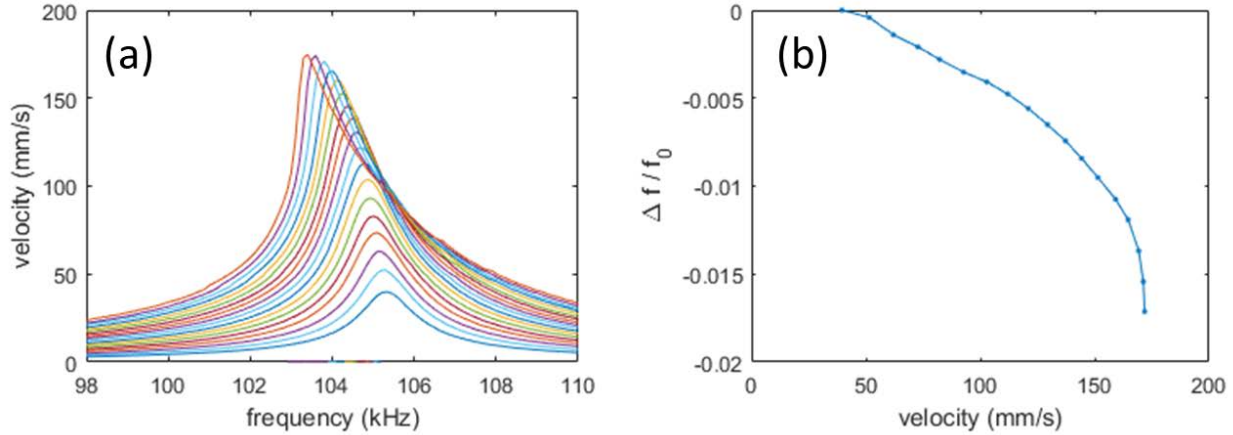


Figure 3. (a) Example resonance curve and (b) corresponding frequency shift plot.

Figure 4 (a) shows the value of α for three different artificial aging conditions of PETN: (a) 30 pristine pellets, (b) 10 pellets aged for 4 weeks at 80°C, and (c) pellets aged for 7 weeks at 80°C. For the pristine pellets, α is tightly centered at approximately 1 (m/s)⁻¹. However, for the 4 week and 7 week pellets, the distribution is greater and ranges from approximately 0 (linear) to 14 (m/s)⁻¹ and 20 (m/s)⁻¹, respectively. While there are aged pellets which have values similar to the pristine sample, from the large spread, it is clear that artificial aging has an effect on the nonlinear acoustic properties of the PETN pellets. The pellets aged for 10 weeks only showed a small increase in the hysteretic nonlinearity as compared to pristine samples, however, three out of the 10 samples were not measureable using NRUS because no isolated peak could be found, compared to 1 sample that was not measureable for the rest of the sample groups as seen in Figure 4 (b).

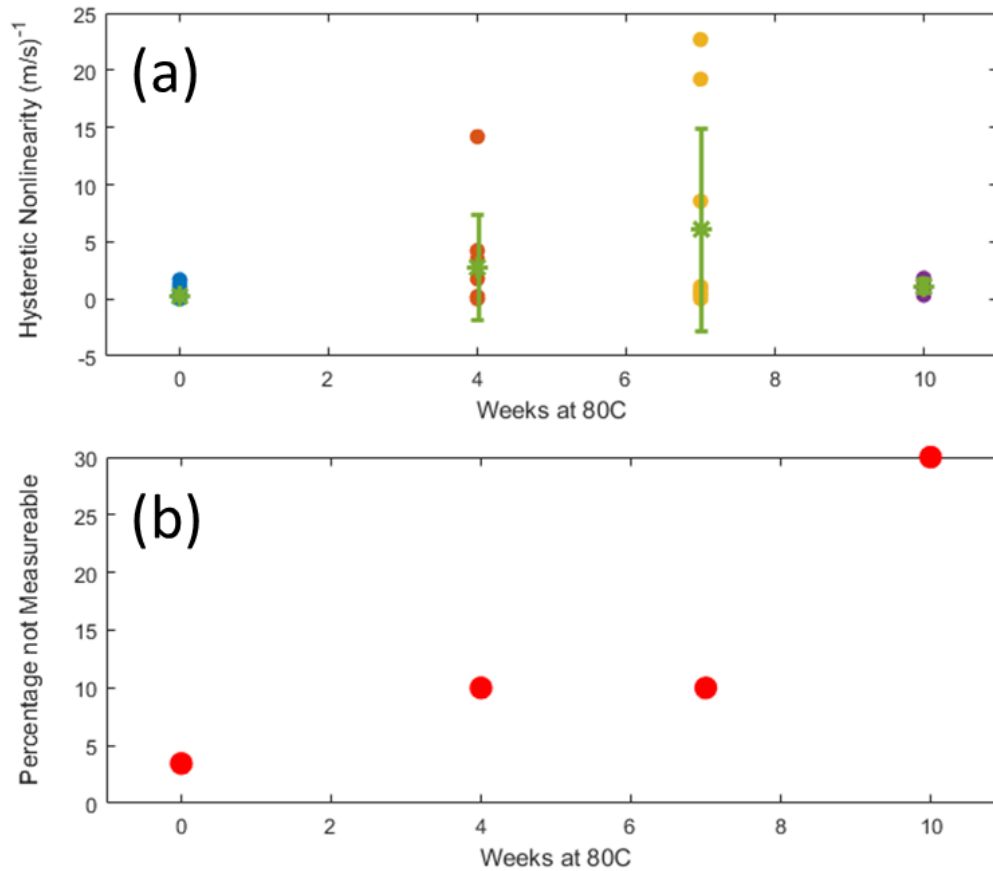


Figure 4. (a) Measured nonlinearity with green symbols representing the average and one standard deviation and (b) percentage of spectral scans that were not measureable using NRUS for different artificial aging conditions.

Anticipated Impact on Mission

Presently, the natural aging processes (only at temperatures that a component is liable to see during its service lifetime) of PETN in detonators have only been studied via artificial aging processes, like heating pellets to unusually high temperatures to accelerate the aging process. Additionally, due to both (1) the lack of assets and (2) the lack of a reliable diagnostic, surveillance of these same components is typically limited to binary response tests; specifically, whether or not the component performs as expected after a lifetime of natural aging. These tests, unfortunately do not give us information about how much the component has aged, nor how much longer the component will continue to perform satisfactorily. As a result, there remains no solid connection between accelerated aging protocols and natural aging. NRUS has promise to address both of these issues: (1) NRUS could feasibly be fielded as a true non-destructive, non-contact diagnostic tool. As a result, most surveillance parts, even those earmarked for destructive testing, could be characterized beforehand without increasing the burden on their already limited availability. (2) Measuring acoustic wave propagation properties in naturally aged PETN would provide target lifetime accumulation metrics that would help us connect

artificial aging to natural aging, i.e.: how much “natural age” is incurred by time at elevated temperature, similarly, how long to age a component at elevated temperature to incur a lifetime of natural aging. In summary, activating an NRUS surveillance diagnostic tool can enrich our understanding of the natural aging processes that occur in PETN and the impact age has on explosives performance.

Conclusion

In this work, we have successfully demonstrated the use of NRUS on small sized PETN pellets, and we have observed differences in the nonlinear hysteretic coefficient, α , for different artificial aging conditions. In the immediate future, we will conduct test fire on the PETN pellets used in this work and compare their firing performance characteristics to the degree of nonlinearity. Additionally, we choose to glue transducers to the pellets for efficiency and simplicity, but this introduces concerns. First, despite developing a precise gluing method, the bond between the transducer and the pellet was not consistent, and the epofix epoxy was absorbed into the pellet. This inconsistency may account for some of the spread seen in the nonlinear parameter. Second, we could not create a nondestructive test of NRUS since the transducers could not be removed. Consequently, the next steps in this work involve developing a noncontact source for ultrasonic excitation.

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